Mobility of Source Zone Heavy Metals and Radionuclides: The Mixed Roles of Fermentative Activity on Fate and Transport of U and Cr

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ABSTRACT:

Various U. S. Department of Energy (DOE) low and medium-level radioactive waste sites contain mixtures of heavy metals, radionuclides and assorted organic materials. Over time, water infiltrates the wastes, and releases metals and radionuclides causing transport into the surrounding environment.

We propose that fermentative microorganisms are active in these sites and may control metal and radionuclide migration from source zones (Figure 1). The following overarching hypothesis will drive our research:

Metals and radionuclides can be mobilized by infiltration of water into waste storage sites. Microbial communities of lignocellulose degrading and fermenting microorganisms present in the subsurface of contaminated DOE sites can significantly impact migration by directly reducing and immobilizing metals and radionuclides while degrading complex organic matter to low molecular weight organic compounds. These low molecular weight organic compounds can increase metal and radionuclide mobility by chelation (i.e., certain organic acids) or decrease mobility by stimulating respiratory metal reducing microorganisms.

The objective of our research is to determine the effect of carbon and energy flow through simulated waste environments on metal and radionuclide migration from waste pits and trenches across the DOE complex. Metals and radionuclides can be mobilized by infiltration of water into waste storage sites. Cellulolytic and non-cellulolytic fermentative microorganisms have been chosen as the focus of this research because their activity is a critical first step that we hypothesize will control subsequent fate and transport in contaminated natural systems. Microbial communities of lignocellulose degrading and fermenting microorganisms present in the subsurface of contaminated DOE sites can significantly impact migration by directly reducing and immobilizing metals and radionuclides while degrading complex organic matter to low molecular weight organic compounds. These low molecular weight organic acids and alcohols can increase metal and radionuclide mobility by chelation (i.e., certain organic acids) or decrease mobility by stimulating respiratory metal reducing microorganisms.

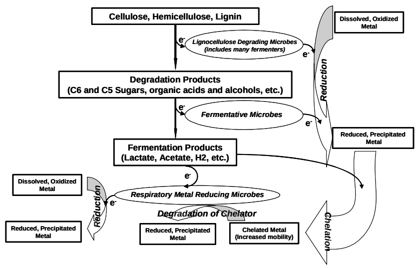

Figure 1. Hypothesized effect of cellulolytic and non-cellulolytic fermentative microbes and their products on metal and radionuclide mobility.

RESEARCH SIGNIFICANCE:

Much of the research performed to understand microbiological reduction of U(VI) and Cr(VI); as well as the effect of low molecular weight organics on metal and radionuclide transport, have only taken into account small segments of the broad spectrum of microbiological activity that occur in the natural environment. While the scientific information produced from this research is highly valuable, identifying the effect of carbon and electron flow through the environment on metal and radionuclide fate would substantially improve predictions of contaminant migration at DOE sites. The proposed research will provide an observational and experimental approach for understanding the coupling of biological, chemical, mineralogical and hydrogeological processes that control fate and transport of U(VI) and Cr(VI) from waste disposal sites at many DOE facilities.

PREVIOUS RESEARCH PROGRESS*:

Our research team has shown:

- Removal of UO_2^{2+} from aqueous solutions using fermenting *Cellulomonas* sp. strain ES6 under anaerobic, non-growth conditions in bicarbonate and PIPES buffer (Figure 2),
- *Cellulomonas* spp. can significantly contribute towards the reduction of not only Cr(VI) and U(VI), but also a variety of iron minerals and dissolved ferric iron sources including hydrous ferric oxide, goethite, maghemite, hematite, magnetite, and Fe(III)-citrate (Figure 3), and
- The presence of quinones increases the electron transport from *Cellulomonas* spp. cells to the iron minerals and also to the dissolved compounds such as Fe(III)-citrate, U(VI), Cr(VI), and TNT (Figures 4-7).

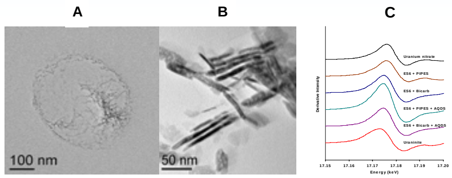

Figure 2. Examination of cultures by transmission electron microscopy (HR-TEM) and energy dispersive X-ray spectroscopy (EDS) showed A) ES6 intracellular U, B) extracellular U accumulation, and C) First derivative of X-ray adsorption near edge structure (XANES) spectra of uranium precipitates indicate that in the presence of *Cellulomonas* sp. ES6 the buffer and the presence or absence of AQDS affected the U(IV):U(VI) ratio.

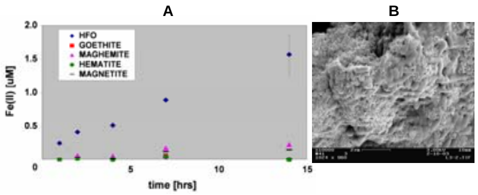

Figure 3. A) Fe(II) production over time from different Fe-minerals by *Cellulomonas* ES6 in the presence of 100 µM AQDS and sucrose. B) *Cellulomonas* sp. strain ES6 on ferrihydrite.

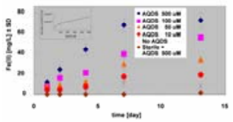

Figure 4. Influence of AQDS on Fe(II) production from hydrous ferrous oxide (HFO) by strain ES6 over time in the presence of sucrose. Inset: Zero order reduction rates of Fe(II) production normalized to cell number.

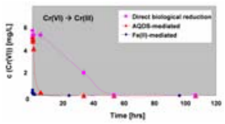

Figure 5. Direct (biological) and indirect (AQDS or Fe(II)-mediated) reduction of Cr(VI) by strain ES6.

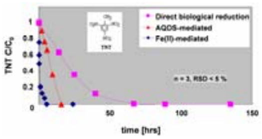

Figure 6. Direct (biological) and indirect (AQDS or Fe(II)-mediated) reduction of TNT by strain ES6.

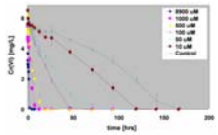

Figure 7. Reduction of Cr(VI) by strain ES6 as a function of AQDS concentration.

PREVIOUS RESEARCH PROGRESS* (Cont):

- Under non-growth conditions *Cellulomonas* sp. ES6 releases phosphate that can readily precipitate U(VI) (Figure 8).

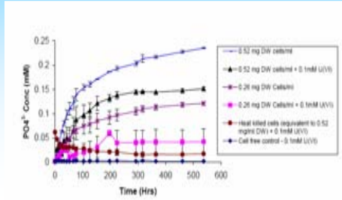

Figure 8. PO_4^{3-} release by ES6 in the presence and absence of U(VI) increases with increasing cell concentration.

FUTURE DIRECTIONS:

Figure 9 provides a schematic summary of research tasks that will be performed as part of the project. Over the next two years our research will focus on:

- Characterizing the production of fermentable substrates and low molecular weight organics from organic debris in low level waste by the activity of cellulolytic and non-cellulolytic fermentative microbial populations and study their effect on the mobility of U(VI) and Cr(VI),
- Understanding the response of microbial communities to carbon and electron flow through these natural and simulated environments, and
- Using this information to develop updated conceptual models for carbon and electron flow in waste systems and the associated effect on Cr(VI) and U(VI) transport in the subsurface.

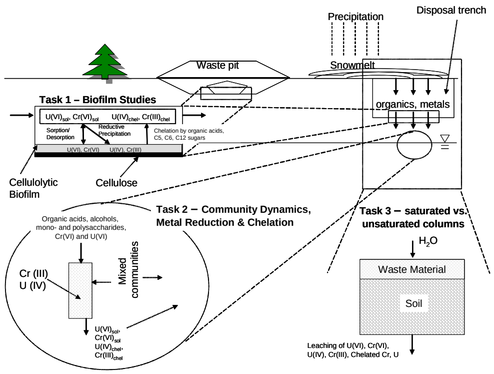

Figure 9. Description of research Tasks and relationship to metal and radionuclide transport from waste pits and trenches into the surrounding environment.

Acknowledgements

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